Atmos. Chem. Phys. Discuss., 11, C5000–C5004, 2011
www.atmos-chem-phys-discuss.net/11/C5000/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Non-linearity in DMS aerosol-cloud-climate interactions" *by* M. A. Thomas et al.

Anonymous Referee #1

Received and published: 16 June 2011

(Anonymous) Referee comment for Thomas, M. A., Suntharalingam, P., Pozzoli, L., Devasthale, A., Kloster, S., Rast, S., Feichter, J., and Lenton, T. M.: Non-linearity in DMS aerosol-cloud-climate interactions, Atmos. Chem. Phys. Discuss., 11, 15227-15253, doi:10.5194/acpd-11-15227-2011, 2011.

General Comments:

Overview: The paper is a short decsription of ECHAM5-HAMMOZ simulations using different DMS emission rates and finding out the CDNC sensititivity. Their main point is that the relationship is non-linear, and they suggest OH limited process to explain this non-linearity. This result would then have implications on DMS-cloud-climate-DMS feedbacks.

I find the article generally well written. The ECHAM5-HAMMOZ is also a good tool for this kind of study and the results should be applicable for other groups as well.

There are some issues related to the overall explanation of the work.

What I found rather surprising, was that the authors were (apparently?) surprised on the fact that the dCDNC/dfDMS rate was not constant, i.e. the CDNC rates did not linearly depend on the increase of the DMS flux rate. I would have instead found any linear dependence very surpsising indeed. This is even more puzzling, as the authors spend relatively large part of the Introduction (pages 15229-15230) to explain previous studies where the non-linearity was already found. As the authors do however study the rate of non-linearity, this is somewhat acceptable. I would maybe revise at least the title to show that non-linearity itself is not the main result, the study concerns more on the rate of non-linearity.

A much more interesting, and potentially more problematic, issue is the non-linearity level itself. If we now consider that the doubling of the DMS will produce CDNC increase of 25% in the region of the interest. However, as there is no comparison to any actual DMS, SO2 or H2SO4 measurements (probably due lack of measurements), there is no way to actually know which part of the non-linearity we are in. If the system truly (in real life, not in model) is OH limited, the actual level of the DMS is critical, not the rate of change. If we do have wrong levels to start with in the model, the non-linearity from OH limited processes might either modelled too high (if DMS levels are lower in real life) or low. This is especially worrying as the actual levels of DMS emissions are not (to my understanding) very well characterized. How do the levels of DMS emissions used in this paper compare to earlier studies?

Thus the point is, the level of non-linearity from OH levels should be strongly dependent on the DMS emissions, and thus only 3 points (0-emission, base case, 2*base case) will only provide quite coarse way to see this. For more general results, we would need

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to know at least 2 more points in between (or higher DMS) to see when the non-linearity starts and how it behaves with DMS increase. This would also serve as a good way to further support (or not) the main hypotheses of the paper (OH limitation). There is an other (weaker) way to study this, as the aerosol/cloud processes generally are much faster than 1/month. One could maybe see the non-linearity already by comparing the monthly averaged DMS fluxes to similarly averaged cloud parameters.

To improve this article for ACP publication, I would recommend answering the following in the manuscript:

1. What kind of situation would create such doubling in DMS fluxes? Is the doubling possible based on the current levels of knowledge? Comparable climate change induced changes are usually in range of +6.5% (Houghton et al, 2001), +2.4% (Bopp et al, 2004) and -8.0% (Stier et al, 2006)..

2. Are the simulated SO2, DMS and H2SO4 levels reasonable compared to (similar, not necessarily from the same region) measurements? Are the DMS emissions used in the base-case simulations reasonable (as far as can be determined)?

3. Are there indications that there is a linear region and non-linear region of the dCDNC/dfDMS or d(CD r)/dfDMS?

4. Are there major sinks of OH not included in the simulations? Such as organic emissions from the oceans? How could they influence the non-linearity?

5. I would also like to see more discussion on other potential non-linearities. Are the overall CDNC, TOA and CD effective radii nonlinearities dominated by the OH? There are many other factors in the the aerosol-cloud processes which could significantly change if the system is perturbed. Examples are: oxidation rates of other pollutants, growth of large particles (resulting in decrease of almost-CCN sized particles), increases in deposition rates of the grown particles, new particle

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formation, etc. These should be more discussed than actually simulated. Do the authors have a good reason to believe the OH process is the dominating one?

6. The DMS emission rates are strongly dependent on the meteorological conditions. In the simulations done in this work, the model was nudged to ECMWF fields. However, the nudging on my understanding will not produce exactly same meteorological fields due normally every 6h nudging. Do the surface wind fields show strong variability between the simulations (probably not)?

7. Do the authors consider other boundary conditions of their work as a major influence of the results? These include resolution, period of the study (2000), location of comparison region (75-30S), relatively old-fashioned new particle formation routine (Vehkamaki, 2002), sea salt emission rates, etc?

Final verdict: Major corrections (because of probably needed new simulations, otherwise ok)

Specific comments:

Regarding tables: Is there a specific reason why in e.g. Table 1, the row title explanation is given above the column title explanation. This is confusing (and the reason for the arrows?) as the column titles are aligned on the top of the table cell. I would either change the order (parameters // diagnostics) or remove the explanations altogether and explain in the figure caption.

Figures have (at least in my PDF) atrocious resolution. They are readable, but should really be given in better format for publication.

References

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 15227, 2011.